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(54) BIODEGRADABLE FILM LAMINATE AND BIODEGRADABLE LABEL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a biodegradable polylactic acid film laminate having a metal thin film layer having metal gloss, and a biodegradable label using the biodegradable film laminate as a support.

SOLUTION: The biodegradable film laminate is constituted by forming the metal thin film layer (A) on a biodegradable film layer (C) through an anchor agent layer (B). The anchor agent layer contains an aliphatic polyester wherein a molar ratio (L/D) of an L-lactic acid residue and a D-lactic acid residue is 1-9, reduced viscosity is 0.3-1.0 dl/g and the concentration of a reactive group is 100-500 equivalent/106 g.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] More specifically, this invention relates to the biodegradability polylactic acid film layered product which prepared the thin film layer which has metallic luster about a biodegradability film layered product. Furthermore, it is related with the biodegradability label, for example, labels, such as a sticker and NEMA of industrial use, using the biodegradability film layered product which has said metallic luster as a base material.

[0002]

[Description of the Prior Art] From the rise of consciousness to an environmental problem in recent years, development of the goods using a natural material or a biodegradability configuration material is performed briskly. The need of the cast using a biodegradable plastic is being extended especially. Various labels aiming at the publication of notes or a trade name, improvement in design nature, etc. are stuck on this front face at the common plastic part in many cases. However, there is a problem of remaining in soil, without disassembling chisels, such as a sticker which consists of plastic resin of non-biodegradability, NEMA, and a label, when the sticker which consists of biodegradability plastic resin, NEMA, a label, etc. are stuck.

[0003] The plastic film which generally has a metal thin film layer for the purpose of improvement in design nature or visibility is used for films for labels, such as NEMA, as a base material in many cases, and the polyethylene terephthalate film with a thickness of 25 to about 200 micrometers is usually used as plastic film. However, the label of biodegradability is proposed in consideration of the above environmental problems (JP,7-44104,A). According to this official report, the polyester system resin which is biodegradability resin as a base material film, naturally-occurring polymers, microorganism production plastics, and polylactic acid system resin are used. It is desirable to use the biodegradation resin of a polylactic acid system as a base material film for labels from fields, such as workability, productivity, various kinetic property, and a printability, as resin of biodegradability.

[0004] However, when fabricating a metal thin film layer on a polylactic acid film and being based on general vacuum deposition, the badness of the adhesion of a polylactic acid film and a metal thin film layer is pointed out. The fault referred to as carrying out interlaminar peeling especially of the biodegradability film which has a metal thin film layer by back processes, such as adhesion processing, printing, a lamination of a protection film, and punching processing, and degradation with time after making it actually stick as a label when using as a base material for labels between a biodegradability film and a metal thin film layer had arisen.

[0005] Conventionally, there is the approach of carrying out surface treatment of the film to a metal thin film layer by sputtering, corona treatment, etc. as a means which raises the adhesion between plastic film. Sputtering is disadvantageous in respect of productivity or cost, and, in the case of a polylactic acid film, the effectiveness of the improvement in adhesion is hardly acquired for corona treatment.

[0006] Moreover, although there is also the approach of using the anchoring agent and 2 liquid hardening mold anchoring agent which consist of polyurethane system resin or polyester system resin as

other means which raise the adhesion between a metal thin film layer and plastic film, when using a polylactic acid film, the effectiveness of the improvement in adhesion cannot be expected.

[0007]

[Problem(s) to be Solved by the Invention] The key objective of this invention is to heighten the layer indirect arrival force and offer [raise the adhesion between a metal thin film layer and plastic film,] the high film layered product of biodegradability, and the biodegradability label using this biodegradability film layered product as a base material further.

[0008]

[Means for Solving the Problem] This invention is the biodegradability film layered product by which the metal vacuum evaporatio no membrane layer (A) was formed in the biodegradability film layer (C) through the support agent layer (B), and said support agent layer is a biodegradability film layered product to which the mole ratio (ratio of length to diameter) of L-lactic acid residue and D-lactic-acid residue is characterized by including the aliphatic series polyester 1-9, and whose reduced viscosity are 0.3 - 1.0 dl/g. As for said aliphatic series polyester, it is desirable to have a reactant radical and a polar group, and 100-500Eq / 106g are suitable for the concentration of these total quantities.

[0009] As for the reactant radical or polar group of said aliphatic series polyester, it is desirable that it is a kind at least among a hydroxyl group, an epoxy group, the amino group, an imino group, a carboxylic-acid radical, a sulfonic group, phosphonic acid radicals, and these salts here. And said aliphatic series polyester has the desirable thing of an isocyanate compound, an epoxy resin, and formaldehyde resins for which the bridge is constructed at least above a kind.

[0010] Furthermore, said biodegradability film is a polylactic acid system film suitably. Other gestalten of this invention are the biodegradability labels which used said biodegradability film layered product as a base material.

[0011]

[Embodiment of the Invention] This invention is the biodegradability film layered product by which the metal thin film layer (A), for example, a metal vacuum evaporatio no membrane layer, was formed in the biodegradability film layer (C) through the support agent layer (B).

[0012] A support agent layer (B) contains the aliphatic series polyester 1-9, and whose reduced viscosity the mole ratios (ratio of length to diameter) of L-lactic acid residue and D-lactic-acid residue are 0.3 - 1.0 dl/g here.

[0013] The support agent layer used for this invention is formed by applying the anchoring agent which dissolved aliphatic series polyester in the non-halogen system organic solvent on a biodegradability film layer.

[0014] By using a lactic acid as a principal component, the aliphatic series polyester used for an anchoring agent carries out a polymerization, and is obtained. and lactic-acid residue -- desirable -- more than 60 mol % -- more -- desirable -- more than 70 mol % -- further -- desirable -- more than 80 mol % - - especially -- desirable -- more than 90 mol % -- most -- desirable -- more than 95 mol % -- it contains. If lactic-acid residue is less than [60 mol %], good bond strength and good biodegradability may not be acquired. Moreover, as for the content of the lactic-acid residue of the above-mentioned aliphatic series polyester, it is desirable that it is less than [99.99 mol %] from an adhesive field.

[0015] Moreover, as for the above-mentioned aliphatic series polyester, both in quality and in quantity, it is preferably desirable to contain lactic-acid residue more than 95 mass % still more preferably more than 90 mass % more preferably more than 80 mass %.

[0016] The above-mentioned aliphatic series polyester can include the polyester block which consists of the copolymer or dicarboxylic acid, and diol of a lactic acid and other oxy acid in a polymer molecule. As oxy acid other than a copolymerizable lactic acid, a glycolic acid, a 2-hydroxy isobutyric acid, 3-hydroxybutyric acid, 16-hydroxy hexadecanoic acid, 2-hydroxy-2-methyl butanoic acid, 12-hydroxy stearic acid, 4-hydroxybutyrate, 10-hydroxy stearic acid, a malic acid, a citric acid, a gluconic acid, etc. are mentioned. Moreover, intramolecular ester of a hydroxy acid like a caprolactone and the cyclic ester which lost and generated the water molecule from alpha-oxy acid like a lactide are also used.

[0017] As dicarboxylic acid, a succinic acid, an adipic acid, an azelaic acid, a sebacic acid, etc. are

mentioned, and ethylene glycol, a diethylene glycol, propylene glycol, butanediol, etc. are mentioned as diol. In addition, calculation of the content (mol %) of the lactic acid at the time of carrying out copolymerization of the polyester which consists of dicarboxylic acid and diol calculates dicarboxylic acid and diol as each unit. Moreover, polyglycerin is also put in and computed to the number count of mols as a polyol component.

[0018] As for the mole ratio (ratio of length to diameter) of the L-lactic acid of the lactic-acid residue in the above-mentioned aliphatic series polyester, and D-lactic acid, it is desirable that it is in the range of 1-9. When D-lactic acid is superfluous when ratio of length to diameter is smaller than 1 namely, it may become high in cost. Moreover, when ratio of length to diameter exceeds 9, the solubility over the general-purpose solvent of a non-halogen system falls, and it may be hard coming to carry out coating of the anchoring agent.

[0019] Although the mole ratio of the L-lactic acid residue in polyester and D-lactic-acid residue was calculated from the charge, it checked that it was the same as what determined the mole ratio of the L-lactic acid residue in polyester, and D-lactic-acid residue using the rotatory-polarization photometer (Horiba SEPA-200).

[0020] With the non-halogen system solvent which dissolves said anchoring agent, ethers solvents, such as ester solvents, such as aromatic solvents, such as ketones, such as a methyl ethyl ketone, methyl isobutyl ketone, and a cyclohexanone, toluene, and a xylene, ethyl acetate, propyl acetate, and butyl acetate, tetrahydrofurane, and diethylether, etc. are mentioned.

[0021] As for the reduced viscosity of the above-mentioned aliphatic series polyester, it is desirable that it is 0.3 - 1.0 dl/g. When reduced viscosity is less than 0.3 dl/g, HAJIKI may arise at the time of coating, or the lack of bond strength may arise. Moreover, when reduced viscosity exceeds 1.0 dl/g, the viscosity of coating liquid may become high and coating fitness may fall.

[0022] In addition, reduced viscosity is 25 degrees C in the sample concentration of 0.125g / 25ml, measurement solvent chloroform, and measurement temperature, and is the value measured using the Ubbelohde-viscosity tubing.

[0023] As for the glass-transition temperature (it is called Following Tg) of the above-mentioned aliphatic series polyester, it is desirable that it is in the range of 35-60 degrees C. When the bond strength of a metal thin film layer may become inadequate when Tg is less than 35 degrees C, and Tg exceeds 60 degrees C, it becomes easy to produce a crack in a metal thin film layer, and may be inferior to design nature or gas barrier property. The desirable minimum of Tg is 38 degrees C, and a still more desirable minimum is 40 degrees C. moreover, here whose a still more desirable upper limit the desirable upper limit of Tg is 57 degrees C, and is 55 degrees C -- Tg -- sample 10mg -- an aluminum pan -- taking -- an aluminum lid -- covering -- strong -- crimp -- carrying out -- this -- DSC (differential scanning calorimeter) -- it is the value measured with 10-degree-C programming rate for /by law.

[0024] As for aliphatic series polyester, it is desirable to have a reactant radical or a polar group, and the range of 100-500Eq / 106g is suitable for the concentration of these sum totals in this case.

[0025] If the sum density of a reactant radical and a polar group is 100Eq / less than 106g, the adhesion of a good metal thin film layer, for example, the metal vacuum evaporation film, will not be acquired, or in the case of bridge formation, it becomes that it is hard to be used for the application as which thermal resistance may fall, without reacting enough with a cross linking agent, and the thermal resistance which is about 50-60 degrees C is required. Moreover, when 500Eq / 106g are exceeded, a water resisting property may make it low.

[0026] A reactant radical is a functional group which can react with other functional groups and can form covalent bond here, for example, a hydroxyl group, an epoxy group, the amino group, an imino group, a carboxylic-acid radical, etc. are mentioned. Moreover, as a polar group, a hydroxyl group, the amino group, an imino group, a carboxylic-acid radical, a sulfonic group, phosphonic acid radicals, and these salts are raised, for example. As said salt, in the case of the amino group and an imino group, it is haloid salt with chlorine, a bromine, etc., or acetate, and, in the case of a carboxylic-acid radical, a sulfonic group, and a phosphonic acid radical, alkali-metal salts, such as a potassium and sodium, ammonium salt, etc. are mentioned.

[0027] It is desirable at the point that adhesion with an expensive hydroxyl group is acquired in these reactant radicals or a polar group. The concentration of these functional groups can be measured by well-known approaches, such as count or titration, from a charge.

[0028] In addition, if how to make a hydroxyl group into an example and ask it for the concentration of a reactant radical is explained The concentration of a hydroxyl group may add and ask for the acid number (the acid number by decomposition of a lactide = hydroxyl value by decomposition of a lactide) which measured the acid number to the value (from the hydroxyl value of polyhydric alcohol or polyglycerin to the calculation added for a reaction) calculated from the charge, and was obtained, and further Superfluous phenyl isocyanate is added, a resin hydroxyl group can be made to be able to react, phenyl isocyanate unreacted next can be made to be able to react with superfluous diethylamine, and it can also ask with the titrimetric method of titrating the amount of unreacted diethylamines with an acid.

[0029] In the case of a hydroxyl group, the approach of adjusting these reactant radicals to a predetermined density range has the following approach. For example, the approach of making react with the approach of adjusting the molecular weight of polyester, the approach of carrying out the polymerization of the polylactic acid using a lactide or the approach of adding a polyhydric-alcohol compound during a polymerization, the approach of adding polyhydric alcohol for polylactic acid after a polymerization, and depolymerizing, a hydroxy group content epoxy compound, etc., and introducing two or more hydroxyl groups in an end etc. is mentioned.

[0030] As polyhydric alcohol, saccharides, such as polyglycerin, polyvinyl alcohol, a sorbitol, a glucose, and a galactose, pentaerythritol, etc. are mentioned. Polyglycerin is desirable also in these.

[0031] As for the polymerization degree of polyglycerin, 3-20 are desirable, it is five or more more preferably, and an upper limit is 15 or less more preferably. The adhesion to a metal thin film layer with polymerization degree good at less than three may not be acquired. Moreover, when the polymerization degree of polyglycerin exceeds 20, a water resisting property may get worse.

[0032] The content of polyglycerin is below 3 mass % especially preferably below 5 mass % still more preferably below 10 mass % more preferably [below 20 mass / in aliphatic series polyester / % is desirable, and]. Moreover, the content of polyglycerin is more than 0.2 mass % especially preferably more than 0.1 mass % still more preferably more than 0.05 mass % preferably more than 0.01 mass [in aliphatic series polyester] %.

[0033] When the content of polyglycerin exceeds 20 mass %, a water resisting property may get worse. Moreover, the adhesive strength of a metal thin film layer may decline under by 0.01 mass %.

[0034] By the approach of introducing the reactant radicals or polar groups other than a hydroxyl group, the approach of carrying out ring opening polymerization of the lactide under existence of the poly allylamine, a poly methacrylic amine, Polly N-ethylamino acrylate, diethanolamine, etc. is mentioned as an example in the case of the amino group. Moreover, as an example in the case of an imino group, the approach of carrying out ring opening polymerization of the lactide is under existence of polyethyleneimine etc. Moreover, as an example in the case of a carboxylic-acid radical, the approach of carrying out ring opening polymerization of the lactide is under existence of the Pori (meta) acrylic acid etc. Furthermore as an example in the case of a sulfonic group, the approach of carrying out ring opening polymerization of the lactide is listed to the bottom of existence of sulfoisophtharate etc.

[0035] In this invention, especially the manufacture approach of said aliphatic series polyester is not limited, but the conventional well-known approach can be used for it. For example, the lactide which is the dimer of a lactic acid, the approach of carrying out melting mixing of other oxy acid etc., and using and carrying out heating ring opening polymerization of the well-known ring-opening-polymerization catalyst (for example, octylic acid tin, aluminium acetylacetonato), the approach heating and reduced pressure perform a direct dehydration polycondensation, etc. are mentioned.

[0036] In this invention, cross linking agents, such as formaldehyde resins, such as polyfunctional isocyanate, polyfunctional epoxy, melamine resin, and benzoguanamine resin, a viscosity controlling agent, a degradation inhibitor, a coloring agent, etc. can be blended with an anchoring agent if needed.

[0037] Especially, the isocyanate of an aliphatic series system is desirable as a cross linking agent. As isocyanate of an aliphatic series system, hexamethylene di-isocyanate, lysine diisocyanate,

hydrogenation toluylene di-isocyanate, hydro-diphenylmethane diisocyanate, etc. are mentioned, and especially these trimers are desirable here.

[0038] Moreover, as for said anchoring agent, it is desirable that the above-mentioned aliphatic series polyester is included especially in a range of more than 90 mass % more than 80 mass % more than 70 mass % in solid content from a viewpoint which promotes biodegradability.

[0039] an anchoring agent -- a biodegradability film layer -- it applies to a polylactic acid film preferably, and a support agent layer is formed. Especially the method of application of an anchoring agent is not limited, but well-known approaches, such as a reverse roll coater, a gravure coating machine, a micro gravure coating machine, a comma coating machine, a bar coating machine, and an air doctor coating machine, are used.

[0040] Although especially the thickness of a support agent layer is not limited, 2.0 micrometers or less are desirable at 0.01 micrometers or more. a crack detailed in a metal thin film layer if the adhesion reinforcement of a film and a metal thin film layer is not fully obtained as the thickness of a support agent layer is less than 0.01 micrometers, but 2.0 micrometers is exceeded, in case a metal thin film layer will be formed with vacuum deposition -- generating -- an exterior -- there is a case where it becomes less desirable. Moreover, when using as a packing material, gas barrier property may fall.

[0041] Moreover, in order to carry out coating of the anchoring agent to a biodegradability film, the in-line coat where coat *Perilla frutescens* (L.) Britton var. *crispa* (Thunb.) Decne. back-extends an anchoring agent before extension of a biodegradability film, or the off-line coat which carries out coating at another process after extension is sufficient.

[0042] The thing of the optical purity beyond 97 mol % has the desirable content of a Poly L-lactic acid acid film, especially L-lactic acid residue, and a biodegradability film layer (C) has a still more desirable thing beyond 99 mol %, although a polylactic acid film is used suitably.

[0043] The laminating of the support agent layer (B) is carried out to a biodegradability film layer (C), and, as for the biodegradability film layered product of this invention, a metal thin film layer is formed on it. And a metal thin film layer is formed in the metal vacuum evaporation layer which has metallic luster preferably. And as a metal vacuum evaporation layer which has metallic luster, although aluminum, gold, silver, copper, chromium, nickel, tin, titanium, etc. and these alloys are mentioned, it is not limited to especially these. Aluminum is mentioned as a cheap metal vacuum evaporation layer. A vacuum deposition method, the sputtering method, etc. are mentioned as the metal vacuum evaporation approach, and it will not be limited if it is the approach of making a metal the shape of the shape of a molecule, or a cluster, and carrying out a laminating on a biodegradability film layer (C).

[0044] As thickness of a metal thin film layer (A), it may be desirable, and 10nm - 100nm cannot acquire metallic luster sufficient in less than 10nm, and may not be desirable as a charge of label material. Moreover, gas barrier property may fall as a packing material, and it may not be desirable. However, it may vapor-deposit to less than 10nm, design nature may be sent as the shape of a half mirror, and especially the thickness of a metal thin film layer is not limited. Moreover, when a metal thin film layer exceeds 100nm, not only the adhesion reinforcement of a film and a metal thin film layer may fall, but there is no merit in cost.

[0045] A biodegradability label can be created using a top Norio resolvability film layered product. An exfoliation sheet is laminated after forming an adhesive layer using the rubber or resin which has the adhesiveness and biodegradability of natural rubber, terpene system resin, or polylactic acid system resin in the metal thin film layer side of a top Norio resolvability film layered product, or the opposite side of a metal thin film layer. Or after applying an adhesive layer beforehand on an exfoliation sheet, you may laminate in the biodegradability film layered product which has metallic luster. Since an exfoliation sheet is what is discarded after use of a label, use of the material which does not give a load preferably to natural environment after abandonment, such as plastics of biodegradability or paper, is desired, but especially in order to use it, dissociating at the time of label use, it may not have biodegradability.

[0046] Furthermore, the biodegradability film layered product, especially polylactic acid film layered product of this invention prepare a heat-sealing layer, apply the use or the binder as packing materials,

such as saccate food, and adhesives, and it can consider as a seal and a label or they can use them as a sticker or a tag.

[0047] Moreover, a printing layer and other coat layers can also be prepared between a biodegradability film layer (C) and support agent layers (B) and in the metal thin film layer (A) and opposite side of a biodegradability film layer (C). Moreover, even if it is the case where a metal thin film layer is formed by the vacuum plating of aluminium by coloring it a support agent layer and other coat layers, it can be made a color tone like golden vacuum evaporation or copper vacuum evaporation, or design nature can be raised by coloring. Especially limitation will not be carried out, if printing ink or a coat layer do not have physical-properties degradation even if it leaves them under natural environment. However, when using the film of a polylactic acid system for a biodegradability film layer, it is desirable from the field of adhesion and biodegradability to use ink and the coating agent of a polylactic acid system. Moreover, in order to raise the adhesion of a biodegradability film layer and a printing layer, an easily-adhesive coat may be established in the front face of a biodegradability film layer. Furthermore, in order to raise the weatherability of a printing layer, and abrasion resistance, a film is also laminable on the front face of a printing layer.

[0048]

[Example] Hereafter, although this invention is explained more to a detail based on an example, it is not limited to these.

(1) polyester (I) be obtained by teach the aluminium acetylacetonato 1 section to 4 opening flask, carry out ring opening polymerization and make a residual lactide distill off under reduced pressure by carry out heating melting at 180 degrees C under nitrogen gas atmosphere mind for 3 hours as the manufacture example ADL lactide 1000 section of aliphatic series polyester, the polyglycerin (die cel chemistry PGL10: hydroxyl group concentration 850 KOHmg/g) 10 section whose polymerization degree be 10, and a ring opening polymerization catalyst.

[0049] In order to ask for the hydroxyl-group concentration of polyester (I), theoretically, the hydroxyl-group concentration of polyester (I) is decided only with the hydroxyl group of the polyglycerin origin. Moreover, the acid number is also set to 0. However, it is common in a raw material that the impurity is contained in fact, and the lactyl lactic acid contained in DL lactide in this case occupies most impurities. In such a case, if the lactyl lactic acid of an impurity works as a polymerization initiator instead of polyglycerin and there is no impurity, the hydroxyl group which should not be produced may arise. Moreover, in the lactyl lactic acid which is oxy acid, since the acid number and a hydroxyl value are equivalence, the hydroxyl-group concentration of the impurity origin can be known by measuring the acid number of polyester (I). Therefore, if the hydroxyl-group concentration of polyglycerin and the measured acid number are applied, the hydroxyl-group concentration of the aliphatic series polyester for which it asks will be obtained.

[0050] Based on the above-mentioned approach, it is the following, and made and asked for the hydroxyl-group concentration of polyester (I). The hydroxyl-group concentration of the glycerol origin in polyester (I) was 150Eq / 106g based on count (KOH conversion hydroxyl-group concentration / mass section of the mass section / polymer of the molecular weight x polyglycerin of KOH) called $(850 \times 1000) / 56 \times 10 / (1000 + 10) = 150$. Moreover, the measured value of the hydroxyl-group concentration of the impurity origin, i.e., the acid number of polyester (I), was 40Eq / 106g. Therefore, the hydroxyl-group concentration of polyester (I) was called for with 190Eq of total value / 106g. In addition, the acid number dissolved (Polyester I) 0.2g in 25ml chloroform, and titrated with the 0.1-N KOH ethanol solution. The phenolphthalein was used as an indicator of titration.

[0051] Polyester (II) was obtained by teaching the aluminium acetylacetonato 1 section to 4 opening flask, carrying out ring opening polymerization and making a residual lactide distill off under reduced pressure by carrying out heating melting at 180 degrees C under nitrogen gas atmosphere mind for 3 hours as the example BDL lactide 1000 section, the polyglycerin (die cel chemistry PGL10: hydroxyl group concentration 850 KOHmg/g) 5.6 section whose polymerization degree is 10, and a ring opening polymerization catalyst.

[0052] Here, when asked for the hydroxyl-group concentration of polyester (II) like the example 1, the

hydroxyl-group concentration of 85Eq / 106g, and the impurity origin of the hydroxyl-group concentration of the polyglycerin origin was 25Eq / 106g. Therefore, the hydroxyl-group concentration of polyester (II) is the total value of 110Eq / 106g.

[0053] Polyester (III) was obtained by teaching the aluminium acetylacetonato 1 section to 4 opening flask, carrying out ring opening polymerization and making a residual lactide distill off under reduced pressure by carrying out heating melting at 180 degrees C under nitrogen-gas-atmosphere mind for 3 hours as the example CDL lactide 1000 section, the polyglycerin (die cel chemistry PGL10: hydroxyl-group concentration 850 KOHmg/g) 16.5 section whose polymerization degree is 10, and a ring-opening-polymerization catalyst.

[0054] Here, when asked for the hydroxyl-group concentration of polyester (III) like the example 1, the hydroxyl-group concentration of 246Eq / 106g, and the impurity origin of the hydroxyl-group concentration of the polyglycerin origin was 30Eq / 106g. Therefore, the hydroxyl-group concentration of polyester (III) is the total value of 276Eq / 106g.

[0055] Polyester (IV) was obtained by teaching the aluminium acetylacetonato 1 section to 4 opening flask, carrying out ring opening polymerization and making a residual lactide distill off under reduced pressure by carrying out heating melting at 180 degrees C under nitrogen gas atmosphere mind for 3 hours as the example DDL lactide 1000 section, the polyglycerin (die cel chemistry PGL10: hydroxyl group concentration 850 KOHmg/g) 26.3 section whose polymerization degree is 10, and a ring opening polymerization catalyst.

[0056] Here, when asked for the hydroxyl-group concentration of polyester (IV) like the example 1, the hydroxyl-group concentration of 409Eq / 106g, and the impurity origin of the hydroxyl-group concentration of the polyglycerin origin was 50Eq / 106g. Therefore, the hydroxyl-group concentration of polyester (IV) is the total value of 459Eq / 106g.

[0057] The presentation and physical properties of polyester (I) - (IV) which were acquired by example A-D are shown in Table 1. Measurement of the specification and physical properties which are shown in Table 1 was based on the following approach.

[0058] It measured using the <reduced viscosity> sample concentration of 0.125g / 25ml, measurement solvent chloroform, the measurement temperature of 25 degrees C, and the Ubbelohde viscosity tubing.

[0059] the point (Tg) transferring [<glass transition point (Tg)> glass point] -- DSC -- it measured by law.

[0060] The lactic-acid residue under <lactic-acid residue (mass %)> lactic-acid system polyester presentation specified the charge, and showed it in Table 1. However, it was checked by 500MHz NMR that the same amount is contained also in polyester.

[0061] Although the mole ratio of the L-lactic acid residue in <mole ratio of L-lactic acid residue and D-lactic-acid residue> polyester and D-lactic-acid residue was calculated from the charge, it checked that it was the same as what determined the mole ratio of the L-lactic acid residue in polyester, and D-lactic-acid residue using the angle-of-rotation meter (Horiba SEPA-200).

[0062]

[Table 1]

アンカー剤のポリエステル組成

	実施例 A	実施例 B	実施例 C	実施例 D
ポリエステル	I	II	III	IV
乳酸残基(重量%)	99.0	99.4	98.4	97.4
L/D(モル比)	1	1	1	1
還元粘度(dl/g)	0.49	0.74	0.50	0.33
水酸基濃度(当量/10 ⁶ g)	190	110	276	459
Tg(°C)	49	49	49	48
ポリ乳酸の重合度	10	10	10	10

[0063] (2) As a manufacture example 1 biodegradability film of a biodegradability film layered product,

by gravure coater, the coating liquid which consists of the polylactic resin (polyester (I)) 1 mass section and the toluene 40 mass section was applied to one side of a polylactic acid film with a thickness of 25 micrometers so that desiccation thickness might be set to 0.02 micrometers, and the support agent layer was formed in it. Subsequently, thickness formed the metal thin film layer which is 45nm with the vacuum deposition of aluminum.

[0064] As an example 2 biodegradability film, by gravure coater, the coating liquid which consists of the polylactic resin (polyester (II)) 1 mass section, the toluene 36 mass section, and the cyclohexanone 4 mass section was applied to one side of a polylactic acid film with a thickness of 25 micrometers so that desiccation thickness might be set to 0.02 micrometers, and the support agent layer was formed in it. Subsequently, thickness formed the metal thin film layer which is 45nm with the vacuum deposition of aluminum.

[0065] As an example 3 biodegradability film, by gravure coater, the coating liquid which consists of the polylactic resin (polyester (I)) 1 mass section, the fat group isocyanate (trade name: duranate TPA-100, Asahi Chemical Industry [Co., Ltd.] Co., Ltd. make) 0.1 mass section, the dibutyltin dilaurate (Tokyo formation Industrial company make) 0.02 mass section, and the toluene 40 mass section was applied to one side of a polylactic acid film with a thickness of 25 micrometers so that desiccation thickness might be set to 0.02 micrometers, and the support agent layer was formed in it. Subsequently, thickness formed the metal thin film layer which is 45nm with the vacuum deposition of aluminum.

[0066] The layered product which has an aluminum thin film like an example 3 was obtained except having used example 4 polylactic resin as polyester (III).

[0067] The layered product which has an aluminum thin film like an example 3 was obtained except having made example 5 polylactic resin into ester (IV).

[0068] As an example of comparison 1 biodegradability film, by gravure coater, the coating liquid which consists of the polyester resin (trade name: made in [Toyobo Product company] Byron 200) 1 mass section, the isocyanate compound (trade name: Coronate L, Japanese Polyurethane company make) 0.1 mass section, the toluene 18 mass section, the methyl-ethyl-ketone 18 mass section, and the cyclohexanone 4 mass section was applied to one side of a polylactic acid film with a thickness of 25 micrometers so that desiccation thickness might be set to 0.02 micrometers, and the support agent layer was formed in it. Subsequently, thickness formed the metal thin film layer which is 45nm with the vacuum deposition of an aluminum layer.

[0069] As an example of comparison 2 biodegradability film, corona treatment was performed on one side of a polylactic acid film with a thickness of 25 micrometers, and the metal thin film layer whose thickness is subsequently 45nm was formed with the vacuum deposition of aluminum.

[0070] Direct thickness formed in one side of a polylactic acid film with a thickness of 25 micrometers the metal thin film layer which is 45nm with the vacuum deposition of aluminum as an example of comparison 3 biodegradability film.

[0071] The performance evaluation of the biodegradability film layered product obtained in the example and the example of a comparison was performed by the following approach.

[0072] a measurement of the adhesion reinforcement of a <adhesion on-the-strength> biodegradability film layer and a metal thin film layer sake -- first -- polyester film with a thickness of 25 micrometers -- adhesives (Oriental Morton, Inc.) company make -- desiccation thickness carried out 5 mass sections mixing of the 100 mass sections and CAT-10 for AD-122 with 3 micrometers -- as -- ***** . After having carried out dry laminate, having created the test piece so that the sample vacuum-plating-of-aluminium side of each biodegradability film layered product might turn into an adhesion side on that front face, and aging at 40 degrees C for 24 hours, this test piece was measured by the exfoliating method 180 degrees. The unit of measured value is g/15mm.

[0073] < peel strength: The sample of a predetermined dimension was created from the biodegradability film layered product obtained in waterproof > example and the example of a comparison, it was immersed in 25-degree C water, this sample was paid 1 hour after picking, and the vacuum-plating-of-aluminium side was ground against the finger. The valuation basis is as follows.

O : a vacuum evaporation layer does not separate.

O : a part of vacuum evaporatio layer separated.

** : Most vacuum evaporatio layers separated.

x : It has arranged so that the vacuum-plating-of-aluminium side of the biodegradability film layered product obtained in the example and the example of a comparison by the hot plate which is <peel strength:thermal-resistance> 80 degree C at which it is only immersion and the vacuum evaporatio layer separated may turn up, and the vacuum evaporatio side was ground against cloth. The valuation basis is as follows.

O : a vacuum evaporatio layer does not separate.

O : a part of vacuum evaporatio layer separated.

** : One half extent of a vacuum evaporatio layer separated.

x : Most vacuum evaporatio layers separated.

[0074]

[Table 2]

		実施例 1	実施例 2	実施例 3	実施例 4	実施例 5	比較例 1	比較例 2	比較例 3
生分解性フィルム積層体	生分解性フィルム 材質	ポリ乳酸	ポリ乳酸	ポリ乳酸	ポリ乳酸	ポリ乳酸	ポリ乳酸	ポリ乳酸	ポリ乳酸
	厚さ(μm)	25	25	25	25	25	25	25	25
	アンカー剤層 材質	ポリエステル (I)	ポリエステル (II)	ポリエステル (I)	ポリエステル (III)	ポリエステル (IV)	樹脂 A	-	-
物性評価	厚さ(μm)	0.02	0.02	0.02	0.02	0.02	0.02	-	-
	金属薄膜層 厚さ(nm)	45	45	45	45	45	45	45	45
	密着強度(g/15mm)	450	400	450	440	460	320	15	30
物性評価	剝離界面	A I // PET	A I // PET	A I // PET	A I // PET	A I // PET	PLC // A I	PLC // A I	PLC // A I
	生分解性	○	○	○	○	○	×	○	○
	耐水性	◎	◎	◎	◎	○	◎	△	△
	耐熱性	△	△	◎	◎	◎	◎	△	△

[0075] The above-mentioned evaluation result is shown in Table 2. In the column of the exfoliation interface of Table 2, an aluminum thin film layer and PET mean a support agent layer (polyester), and, as for PLC, aluminum means a biodegradability film layer (polylactic acid film layer).

[0076] All the test pieces of an example exfoliated from Table 2 in the vacuum-plating-of-aluminium layer and the polyester film layer about the evaluation result of peel strength to all the test pieces of the example of a comparison having exfoliated in the vacuum-plating-of-aluminium layer and the biodegradability film layer (polylactic acid film layer).

[0077] Evaluation of biodegradability was performed by the following approach. The test piece of a predetermined dimension is created from the biodegradability film layered product obtained in the example and the example of a comparison. An adhesive layer is formed in the vacuum-plating-of-aluminium side of a test piece by the comma coating machine so that the mass after desiccation may be set to 30g/m² in the mixture of natural rubber and terpene system resin. On the other hand, the releasing paper which prepared the silicone layer in one side is laminated so that said adhesive layer and silicone layer may be put together. Furthermore a printing layer is formed in the front face of a polylactic acid film (biodegradability film), and a biodegradability label is created. After not exfoliating in the interface of a polylactic acid film layer and an aluminum vacuum evaporatio layer in the process which creates a biodegradability label and removing a releasing paper for said biodegradability label further, it stuck on the injection-molded product of biodegradability resin, and it laid under a depth of 10cm among soil.

[0078] Six months after except for the example 1 of a comparison, as for an example and other examples of a comparison, decomposition was checked for the label and the injection-molded product.

[0079] It should be thought that all of the gestalt and example of the operation indicated this time are

instantiation, and they are not restrictive. The range of this invention is shown by the above-mentioned not explanation but claim, and it is meant that all modification in a claim, equal semantics, and within the limits is included.

[0080]

[Effect of the Invention] The biodegradability laminated film layered product which carried out the laminating of the metal thin film layer to the biodegradability laminated film through the support agent layer of biodegradability polyester, especially polylactic resin shows the adhesion reinforcement which was excellent between the layers of a metal thin film layer and a biodegradability film layer as explained above. Moreover, especially the biodegradability label created using this biodegradation laminated film ***** is very useful when used with the injection-molded product which is excellent in design nature and recognition nature, and does not give a load to natural environment and which especially consists of a biodegradable plastic.

[Translation done.]

* NOTICES *

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- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] Said support agent layer is a biodegradability film layered product to which it is characterized by being the biodegradability film layered product by which the metal thin film layer (A) was formed in the biodegradability film layer (C) through the support agent layer (B), and 1-9, and reduced viscosity containing the aliphatic series polyester whose mole ratio (ratio of length to diameter) of L-lactic acid residue and D-lactic-acid residue is 0.3 - 1.0 dl/g.

[Claim 2] Aliphatic series polyester is a biodegradability film layered product according to claim 1 which has a reactant radical and a polar group and is characterized by the concentration of these total quantities being 100-500Eq / 106g.

[Claim 3] The biodegradability film layered product according to claim 2 to which a reactant radical or a polar group is characterized by being a kind at least of a hydroxyl group, an epoxy group, the amino group, an imino group, a carboxylic-acid radical, a sulfonic group, phosphonic acid radicals, and these salts.

[Claim 4] The biodegradability film layered product according to claim 1 to 3 to which aliphatic series polyester is characterized by the thing of an isocyanate compound, an epoxy resin, and formaldehyde resins for which the bridge is constructed at least above a kind.

[Claim 5] A biodegradability film layer is a biodegradability film layered product according to claim 1 to 4 characterized by consisting of polylactic acid system films.

[Claim 6] The biodegradability label using the biodegradability film layered product according to claim 1 to 5 as a base material.

[Translation done.]